Pure spin photocurrents

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The pure spin currents, i.e., the counterflow of particles with opposite spin orientations, can be optically injected in semiconductors. Here, we develop a phenomenological theory, which describes the polarization dependencies of spin currents excited by linearly polarized light in bulk semiconductors and quantum well structures of various symmetries. We present microscopic descriptions of the pure spin photocurrents for interband optical transitions in undoped quantum wells as well as for direct intersubband and indirect intrasubband (Drude-like) transitions in n-doped quantum well structures. We also demonstrate that pure spin currents can be generated in structures of sufficiently low symmetries by simple electron gas heating. The theoretical results are compared with recent experimental observations.

I. INTRODUCTION

By definition, the pure spin current of free carriers, electrons or holes, is a spin flux without an electric current. It can be conceived as formed by opposing equivalent flows of spin-up and spin-down particles. This nonequilibrium distribution of carriers in the wave vector and spin spaces is characterized by zero charge current, because electric currents contributed by spin-up and spin-down particles cancel each other, but leads to the accumulation of opposite spins at the opposite edges of the sample. In particular, a pure spin current is produced in a direction perpendicular to an applied electric field due to spin-dependent skew or side-jump scattering of electrons by impurities or phonons. This effect known as the spin Hall effect was predicted in early theoretical studies [1, 2] and observed in recent years, see [3, 4, 5].

In this study, we present the theory of pure spin currents generated under the light absorption in an unbiased semiconductor structures. In spin-dependent optical spectroscopy, the efforts were mostly directed towards (i) the photogeneration of nonequilibrium spin polarization of carriers, the effect known as the optical orientation (see, e.g., [6, 7]), and (ii) the generation of spin-sensitive electric currents known as the circular photogalvanic, spin-galvanic and magneto-gyrotropic effects [7, 8]. In contrast, the free carriers participating in a pure spin current neither have a net spin polarization nor produce a net charge current. A spacial separation of electron spins caused by the spin photocurrent was first observed by using two-color optical coherence control techniques, due to quantum interference of one- and two-photon absorption of two orthogonally-polarized overlapping laser pulses with frequencies ω and 2ω [9, 10, 11]. Then, Bhat et al. [12] and Tarasenko and Ivchenko [13] showed that merely one-photon absorption of linearly polarized light should produce pure spin currents in noncentrosymmetric bulk semiconductors and quantum well (QW) structures: in this case there is no net motion of charge but spin-up and spin-down photoelectrons travel in the opposite directions. The theoretical prediction was followed by an observation of pure spin currents induced by a single linearly polarized optical pulse in (110)-oriented GaAs QWs [14].

Theoretically, the two-color generation and control of spin currents have been extensively analyzed [12, 15, 16, 17]. Here we will consider, in order, the one-photon generation of pure spin currents in unbiased structures under interband, intersubband and intraband absorption of linearly polarized or unpolarized light and derive equations for the corresponding currents. Particularly, we compare different mechanisms of pure spin currents and show difference in the behavior of their contributions as a function of the light frequency and the polarization direction with regard to the crystallographic axes. We also show that pure spin currents emerge in QW structures as soon as the electron gas is simply driven out of thermal equilibrium with the crystal lattice. Finally, we will discuss a new phenomenon which can be called the pure valley-orbit current and observed in many-valley semiconductors. The role of spin-up and spin-down states in pure spin currents is replaced in the valley-orbit current by the index of the conduction-band valleys: the valleys are equally populated, there is no net charge current, but the electrons in different valleys travel in different directions.

II. PHENOMENOLOGY

Phenomenologically, the spin flux, or, in general, the flux of angular momentum, is described by a second-rank pseudotensor $\mathbf{J}$ whose components J_β^α stand for the flow in the β direction of spins oriented along α, with α and β being the Cartesian coordinates. Nonzero components of the photo-induced spin current $\mathbf{J}$ are determined by the light polarization and the explicit form of spin-orbit interaction governed by the structure symmetry. They can be revealed from the symmetry analysis which requires no knowledge about microscopical mechanisms of the spin current generation. Indeed, in the regime of linear dependence of $\mathbf{J}$ on the light intensity I, the spin-photocurrent components are phenomenologically related by

$$J_\beta^\alpha = I \sum_{\gamma\delta} Q_{\alpha\beta\gamma\delta}\, e_\gamma e_\delta^* \,, \qquad (1)$$

to the light-polarization unit vector $\mathbf{e}$ and the complex conjugate vector $\mathbf{e}^*$. Equation (1) represents the most

general form of the spin photocurrent description because the set of quadratic terms $e_\gamma e_\delta^*$ fully determines the light polarization state [18].

Equation (1) can be usefully rewritten in the equivalent form

$$J_\beta^\alpha = I\sum_{\gamma\delta} L_{\alpha\beta\gamma\delta}\frac{e_\gamma e_\delta^* + e_\delta e_\gamma^*}{2} + I\sum_\gamma C_{\alpha\beta\mu}\mathrm{i}[\mathbf{e}\times\mathbf{e}^*]_\mu, \quad (2)$$

where $L_{\alpha\beta\gamma\delta} = (Q_{\alpha\beta\gamma\delta} + Q_{\alpha\beta\delta\gamma})/2$ is a fourth-rank pseudotensor symmetric in the two last indices, $C_{\alpha\beta\mu} = \sum_{\gamma\delta} Q_{\alpha\beta\gamma\delta}\,\epsilon_{\gamma\delta\mu}/(2\mathrm{i})$ is a third-rank tensor, and $\epsilon_{\gamma\delta\mu}$ is the completely antisymmetric third-rank pseudotensor, or the Levi-Civita tensor. The pseudotensor $\mathbf{L}$ describes spin photocurrents which are independent of the sign of light circular polarization for elliptically polarized light and can be conveniently measured for the linearly polarized radiation. In contrast, the tensor $\mathbf{C}$ stands for helicity-sensitive spin photocurrents which reverse their polarity upon switching the sign of circular polarization. This occurs because the cross product $\mathrm{i}[\mathbf{e}\times\mathbf{e}^*]$ is zero for linearly polarized light and proportional to the light helicity for elliptical or circular polarization. Usually, the absorption of circularly polarized light results in a considerable spin polarization of photoexcited carriers [6] masking the observation of pure spin currents. Therefore, in what follows, we focus on spin currents excited by linearly polarized light only and assume the polarization vector $\mathbf{e}$ to be real.

In crystals having a zinc-blende structure and characterized by the symmetry point group T_d, the linearly polarized light can induce both diagonal J_α^α and off-diagonal J_β^α ($\alpha \neq \beta$) components of the spin current. Their polarization properties are described by

$$\begin{aligned} J_\alpha^\alpha &= L_1\, I(e_{\alpha+1}^2 - e_{\alpha+2}^2)\,, \\ J_{\alpha+1}^\alpha &= -J_\alpha^{\alpha+1} = L_2 I\, e_\alpha e_{\alpha+1}\,. \end{aligned} \quad (3)$$

Here, I is the light intensity, the index α runs over the cubic axes $x\|[100]$, $y\|[010]$, and $z\|[001]$, and the index $\alpha+1$ is obtained by the cyclic permutation of the indices x, y, z. Note that nonzero values of the phenomenological parameters L_1 and L_2 in Eq. (3) are allowed in noncentrosymmetric crystals of the T_d symmetry and forbidden for diamond-type centrosymmetric crystals.

The symmetry of (001)-oriented QWs grown from zinc-blende-type semiconductors reduce to the point group D_{2d} in symmetrical structures and C_{2v} in asymmetrical structures. For the latter, the spin photocurrent components photoinduced in the (xy) plane are described by 10 linearly independent constants as follows

$$\begin{aligned} J_x^x/I &= L_1^{\mathrm{B}} e_x^2 + L_2^{\mathrm{B}} e_y^2 + L_3^{\mathrm{B}} e_z^2 + L_1^{\mathrm{S}} e_x e_y\,, \\ J_y^x/I &= L_2^{\mathrm{S}} e_x^2 + L_3^{\mathrm{S}} e_y^2 + L_4^{\mathrm{S}} e_z^2 + L_4^{\mathrm{B}} e_x e_y\,, \\ J_x^y/I &= -L_3^{\mathrm{S}} e_x^2 - L_2^{\mathrm{S}} e_y^2 - L_4^{\mathrm{S}} e_z^2 - L_4^{\mathrm{B}} e_x e_y\,, \\ J_y^y/I &= -L_2^{\mathrm{B}} e_x^2 - L_1^{\mathrm{B}} e_y^2 - L_3^{\mathrm{B}} e_z^2 - L_1^{\mathrm{S}} e_x e_y\,, \\ J_x^z/I &= L_5^{\mathrm{B}} e_x e_z + L_5^{\mathrm{S}} e_y e_z\,, \\ J_y^z/I &= -L_5^{\mathrm{S}} e_x e_z - L_5^{\mathrm{B}} e_y e_z\,. \end{aligned} \quad (4)$$

Here, the superscript B marks those coefficients which are allowed in QWs of the D_{2d} symmetry and can be related to bulk inversion asymmetry (BIA) of the host crystal and/or anisotropy of the chemical bonds at the QW interfaces, while the superscript S marks the contributions which appear because of structure inversion asymmetry (SIA) only. Therefore, in symmetrical (001)-grown QWs, the coefficients L_i^{S} vanish and the polarization dependencies of spin current components are completely determined by the terms proportional to L_i^{B}. In the opposite limit, where the SIA predominates and the QW structure can effectively be described by the axial point group $C_{\infty v}$, the spin photocurrent is given by Eq. (4) with the BIA-related terms being disregarded and the coefficients L_i^{S} satisfying the relation $L_3^{\mathrm{S}} = L_1^{\mathrm{S}} + L_2^{\mathrm{S}}$.

It follows from Eq. (4) that, under normal incidence on a (001)-grown QW, the linearly polarized light can excite fluxes of electron spins oriented only in the interface plane. To create the J_x^z and J_y^z spin current components, which can cause the spacial profile of the spin density S_z, one has to irradiate the QW in the oblique-incidence geometry. This is in contrast to QWs grown along low-symmetry crystallographic axes, where the normally-incident light can induce fluxes of both the in-plane and out-of-plane components of the spin polarization.

As an example of such low-symmetry structures, we consider QWs grown on (110)-oriented substrates and use the (x', y', z') coordinate frame with z' along the growth direction and the in-plane axes $x'\|[1\bar{1}0]$ and $y'\|[00\bar{1}]$. Asymmetrically-grown (110)-oriented QWs have the point group C_s and contain only two symmetry elements: the identity and a mirror plane $m_1 = (1\bar{1}0)$ perpendicular to the x' axis. In this particular case, components of the spin current excited by normally-incident light are phenomenologically given by

$$\begin{aligned} &J_{x'}^{x'}/I = L_1' e_{x'} e_{y'}\,, \quad J_{y'}^{x'}/I = L_2' + L_3'(e_{x'}^2 - e_{y'}^2)\,, \\ &J_{x'}^{y'}/I = L_4' + L_5'(e_{x'}^2 - e_{y'}^2)\,, \quad J_{y'}^{y'}/I = L_6' e_{x'} e_{y'}\,, \\ &J_{x'}^{z'}/I = L_7' + L_8'(e_{x'}^2 - e_{y'}^2)\,, \quad J_{y'}^{z'}/I = L_9' e_{x'} e_{y'}\,. \end{aligned} \quad (5)$$

Symmetrical (110)-grown QWs contain an additional mirror plane $m_2 = (110)$ perpendicular to the z' axis. Reflection by the plane m_2 changes the sign of the $J_\beta^{x'}$ and $J_\beta^{y'}$ ($\beta = x', y'$) components of the spin current but does not modify $J_\beta^{z'}$ as well as the in-plane components of the polarization vector $\mathbf{e}$. Therefore, in symmetrical (110)-grown QWs, the parameters $L_1'...L_6'$ vanish and the spin photocurrent is solely described by the last line of Eq. (5).

Microscopically, the emergence of a pure spin current under the light absorption is related to spin-orbit interaction coupling spin states and spatial motion of charge carriers, the latter being directly affected by the electric field of the light. In terms of the kinetic theory, the J_β^α component of the spin photocurrent in the conduction band is contributed by a non-equilibrium correction

$\propto \sigma_\alpha k_\beta$ to the electron spin density matrix, where σ_α is the Pauli matrix and $\mathbf{k}$ is the wave vector. In general, the concept of spin currents is uncertain in systems with spin-orbit interaction, since the spin and spin-dependent velocity cannot be determined simultaneously (see, e.g., Ref. [19]). Mathematically it is caused by the fact that the Pauli matrices and the velocity operator do not commute. However, this problem of the spin current definition emerges in high orders in the spin-orbit interaction only and vanishes for the cases, where spin currents are directly proportional to the constant of spin-orbit coupling. To the first order in the spin-orbit coupling and within the relaxation time approximation, components of the pure spin current photoinduced in the conduction band are given by

$$J_\beta^\alpha = \sum_{\mathbf{k}} \tau_e \mathrm{Tr}\left[\frac{\sigma_\alpha}{2}\, v_\beta(\mathbf{k})\, G(\mathbf{k})\right] , \qquad (6)$$

with the spin-dependent corrections being taken into account either in the velocity operator $\mathbf{v}(\mathbf{k})$ or in the photogeneration rate of the spin density matrix $G(\mathbf{k})$. Here, τ_e is the relaxation time of the spin current which can differ from the conventional momentum relaxation time that governs the electron mobility. Electron-electron collisions between particles of opposite spins, which do not affect the mobility, contribute to the relaxation of pure spin currents reducing the time τ_e (see, the spin Coulomb drag [20, 21] and the effect of electron-electron interaction on spin relaxation [22], and references therein).

III. INTERBAND TRANSITIONS IN QWS

Among microscopic mechanisms of the pure spin photocurrent we first discuss that related to **k**-linear spin-orbit splitting of quantum subbands [13], in the following the split-subband-related mechanism. The mechanism is most easily conceivable for direct transitions between the heavy-hole valence subband $hh1$ and conduction subband $e1$ in (110)-grown QWs. In such structures, the spin component along the QW normal z' is coupled with the in-plane electron wave vector due to the terms proportional to $\sigma_{z'}k_{x'}$ and $J_{z'}k_{x'}$ in the conduction and valence bands, respectively, where $J_{z'}$ is the 4×4 matrix of the angular momentum 3/2 [7]. This leads to **k**-linear spin splitting of both the electron subband $e1$ and the valence subband $hh1$ into branches with the spin projection $\pm 1/2$ and $\pm 3/2$, respectively, as sketched in Fig. 1(a). The corresponding dispersions in the subbands at small in-plane wave vector are given by

$$E^{(e1)}_{\mathbf{k},\pm 1/2} = \frac{\hbar^2(k_{x'}^2 + k_{y'}^2)}{2m_e} \pm \gamma^{(e1)}_{z'x'} k_{x'} , \qquad (7)$$

$$E^{(hh1)}_{\mathbf{k},\pm 3/2} = -\frac{\hbar^2(k_{x'}^2 + k_{y'}^2)}{2m_h} \pm \gamma^{(hh1)}_{z'x'} k_{x'} ,$$

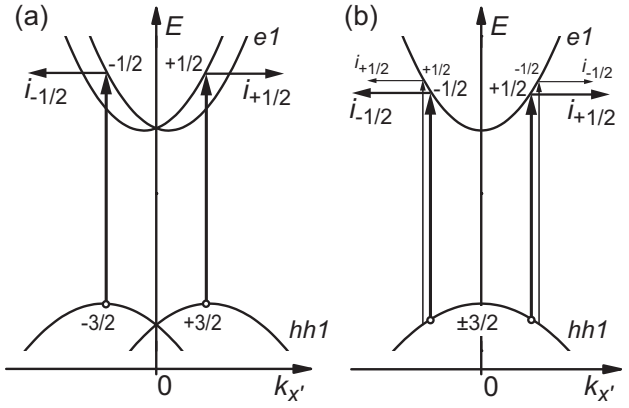

FIG. 1: Microscopic mechanisms of the pure spin photocurrent induced by interband excitation with linearly polarized light in (110)-QWs due to (a) **k**-linear spin splitting of subbands and (b) **k**-linear terms in the transition rates.

where m_e and m_h are the electron and hole effective masses in the QW plane. Note that the spin splitting of the conduction subband is relativistic and, therefore, small as compared to the nonrelativistic term $J_{z'}k_{x'}$ describing the splitting of heavy-hole states in (110)-grown structures.

Due to the selection rules, the allowed direct optical transitions from the valence subband $hh1$ to the conduction subband $e1$ are $|+3/2\rangle \to |+1/2\rangle$ and $|-3/2\rangle \to |-1/2\rangle$ [6], as illustrated in Fig. 1(a) by vertical lines. Under excitation with linearly polarized or unpolarized light the rates of both transitions are equal. In the presence of spin splitting, the optical transitions induced by photons of the fixed energy $\hbar\omega$ occur in the opposite points of the $\mathbf{k}$ space for the spin branches $s_{z'} = \pm 1/2$. Such an asymmetry of photoexcitation results in a flow of electrons within each spin branch. The corresponding fluxes $\mathbf{i}_{+1/2}$ and $\mathbf{i}_{-1/2}$ are of equal strengths but of opposite directions. Thus, this non-equilibrium electron distribution is characterized by the nonzero spin current $(1/2)(\mathbf{i}_{+1/2} - \mathbf{i}_{-1/2})$ but a vanishing charge current, $e(\mathbf{i}_{+1/2} + \mathbf{i}_{-1/2}) = 0$.

To calculate the spin current, we note that the points of optical transitions in the $\mathbf{k}$ space are determined by the energy and quasi-momentum conservation which reads

$$E_g^{QW} + \frac{\hbar^2(k_{x'}^2 + k_{y'}^2)}{2\mu} + 2s_{z'}(\gamma^{(e1)}_{z'x'} - \gamma^{(hh1)}_{z'x'})k_{x'} = \hbar\omega, \quad (8)$$

where E_g^{QW} is the QW band gap at $\mathbf{k} = 0$ and $\mu = m_e m_h/(m_e + m_h)$ is the reduced effective mass. Owing to spin splitting of both the $e1$ and $hh1$ subbands, electrons are photoexcited into the spin branches $s_{z'} = \pm 1/2$ with the average velocities

$$\langle v_{x'} \rangle = \frac{\hbar}{m_e}\langle k_{x'} \rangle + 2s_{z'}\frac{\gamma^{(e1)}_{z'x'}}{\hbar} = 2s_{z'}\frac{\mu}{\hbar}\left(\frac{\gamma^{(e1)}_{z'x'}}{m_h} + \frac{\gamma^{(hh1)}_{z'x'}}{m_e}\right). \qquad (9)$$

The opposite motion of spin-up and spin-down electrons decays within the relaxation time τ_e. However, under the steady-state excitation the electron generation is continuous resulting in the spin current

$$J_{x'}^{z'} = \frac{\mu\tau_e}{2\hbar}\left(\frac{\gamma_{z'x'}^{(e1)}}{m_h} + \frac{\gamma_{z'x'}^{(hh1)}}{m_e}\right)\frac{\eta_{cv}}{\hbar\omega} I\,, \tag{10}$$

where η_{cv} is the QW absorbance.

Another contribution to the spin photocurrent may come from **k**-linear terms in the matrix elements of the interband optical transitions [23], hereafter referred to as the matrix-element-related mechanism. Taking into account $\mathbf{k}\cdot\mathbf{p}$ admixture of the remote conduction band Γ_{15}^c to the valence-band and conduction-band states $X_\mathbf{k}, Y_\mathbf{k}, Z_\mathbf{k}$, and $S_\mathbf{k}$, respectively, one derives the interband matrix elements of the velocity operator for bulk zinc-blende-type semiconductors [24, 25]

$$\begin{aligned} i\langle S_\mathbf{k}|\mathbf{e}\cdot\mathbf{v}|X_\mathbf{k}\rangle &= (P/\hbar)[e_x + i\beta(e_y k_z + e_z k_y)]\,, \\ i\langle S_\mathbf{k}|\mathbf{e}\cdot\mathbf{v}|Y_\mathbf{k}\rangle &= (P/\hbar)[e_y + i\beta(e_x k_z + e_z k_x)]\,, \\ i\langle S_\mathbf{k}|\mathbf{e}\cdot\mathbf{v}|Z_\mathbf{k}\rangle &= (P/\hbar)[e_z + i\beta(e_x k_y + e_y k_x)]\,, \end{aligned} \tag{11}$$

where $P = i(\hbar/m_0)\langle S|p_z|Z\rangle$, $P' = i(\hbar/m_0)\langle S|p_z|Z'\rangle$ and $Q = i(\hbar/m_0)\langle X'|p_y|Z\rangle$ are the interband matrix elements at the Γ point of the Brillouin zone, X', Y', Z' are the Bloch functions of the Γ_{15}^c band, m_0 is the free electron mass, $\beta = QP'(2E_g' + E_g)/[PE_g'(E_g' + E_g)]$ is a material parameter, E_g is the fundamental band gap, and E_g' is the energy separation between conduction bands Γ_{15}^c and Γ_6 at the Γ point. For GaAs, the coefficient β can be estimated as $0.2 \div 1$ Å depending on the band parameters used [26, 27].

The **k**-linear terms in Eq. (11) do not modify the selection rules for optical transitions from the heavy-hole valence subband to the conduction band. As before, the only allowed transitions are $|+3/2\rangle \to |+1/2\rangle$ and $|-3/2\rangle \to |-1/2\rangle$. However, the rates of the above transitions become dependent of the in-plane wave vector. Particularly, for the linearly polarized light normally incident upon a (110)-grown QW, the squared moduli of the matrix elements, which determine the optical transition rates, assume the following form in linear-in-β approximation

$$\begin{aligned} |\langle +1/2|\mathbf{e}\cdot\mathbf{v}|+3/2\rangle|^2 &= P^2/(2\hbar^2) \\ &\times[1 + 2\beta k_{y'} e_{x'} e_{y'} - 2\beta k_{x'}(e_{x'}^2 - e_{y'}^2)]\,, \\ |\langle -1/2|\mathbf{e}\cdot\mathbf{v}|-3/2\rangle|^2 &= P^2/(2\hbar^2) \\ &\times[1 - 2\beta k_{y'} e_{x'} e_{y'} + 2\beta k_{x'}(e_{x'}^2 - e_{y'}^2)]\,. \end{aligned} \tag{12}$$

It follows from Eq. (12) that, for a fixed light polarization, the spin-up and spin-down electrons are predominantly photoexcited in opposite points in the **k** space. This is illustrated in Fig. 1(b) for the light polarized along the y' axis, where electrons with the spin $+1/2$ are generated at a higher rate into states with positive values of $k_{x'}$ whereas electrons with the spin $-1/2$ are mainly generated into states with $k_{x'} < 0$. The difference in rates is shown by vertical lines of different thicknesses. We note that here the spin-orbit splitting of the subbands $hh1$ and $e1$ is unimportant and, therefore, not shown in Fig. 1(b) for simplicity. The spin-dependent asymmetry of optical excitation leads also to the pure spin current. Calculation shows that, in (110)-grown QWs, components of the spin photocurrent caused by **k**-linear terms in the matrix elements of optical transitions have the form

$$J_{x'}^{z'} = \beta(e_{y'}^2 - e_{x'}^2)\frac{\tau_e\varepsilon}{\hbar}\frac{\eta_{cv}}{\hbar\omega} I\,, \quad J_{y'}^{z'} = \beta e_{x'} e_{y'}\frac{\tau_e\varepsilon}{\hbar}\frac{\eta_{cv}}{\hbar\omega} I\,, \tag{13}$$

where $\varepsilon = (\hbar\omega - E_g^{QW})\mu/m_e$ is the kinetic energy of photoexcited electrons. In contrast to Eq. (1), this contribution depends on the polarization plane of the incident light and vanishes for unpolarized light. From comparison of Eqs. (10) and (13) one can see that, depending on the value of $\hbar\omega - E_g^{QW}$, the two contributions to $J_{x'}^{z'}$ can be comparable or one of them can dominate over the other. We also note that the both spin current contributions are caused by bulk inversion asymmetry and do not vanish in symmetrically-grown (110)-QWs.

In (001)-grown QWs, the absorption of linearly polarized or unpolarized light results in a in-plane flow of electron spins, see Eq. (4). In contrast to the low-symmetry QWs considered above, in the (001)-QW structures the linear-in-**k** terms in the matrix elements of optical transitions from the heavy-hole subband vanish at the normal incidence. Since, in addition, the **k**-linear spin splitting of the heave-hole subband is suppressed in (001)-grown structures [7, 28], we conclude that the spin photocurrents are entirely related to spin-orbit splitting of the conduction subband. Assuming the parabolic spin-independent dispersion in the $hh1$ subband and taking into account the spin-dependent contribution

$$H_{\rm so}^{(e1)} = \sum_{\alpha\beta}\gamma_{\alpha\beta}^{(e1)}\,\sigma_\alpha k_\beta \tag{14}$$

to the electron effective Hamiltonian in the subband $e1$, the components of pure spin current generated in the subband $e1$ are derived to be

$$J_\beta^\alpha = \gamma_{\alpha\beta}^{(e1)}\frac{\tau_e\,\mu}{2\hbar m_h}\frac{\eta_{cv}}{\hbar\omega} I\,. \tag{15}$$

For the interband transitions from the light-hole subband or the spin-split band Γ_7, both the split-subband-related and the matrix-element-related mechanisms lead to polarization-dependent pure spin photocurrents. The analysis shows that, in the geometry of normal incidence, the optical excitation from the light-hole subband in (001)-grown QWs leads to the spin current described by Eq. (4) where the phenomenological coefficients satisfy the relations $L_4^B = L_2^B - L_1^B$ and $L_1^S = L_3^S - L_2^S$. If the spin photocurrent is solely caused by **k**-linear terms in the matrix elements of optical transitions then, in addition, $L_1^B = 0$ and $L_2^S = L_3^S = 0$. In the opposite case, when the spin current is mainly contributed by the split-subband-related mechanism, the coefficients are interconnected by

$L_1^B = \pm L_2^B$ and $L_2^S = \pm L_3^S$ with the sign "+" or "−" depending on, respectively, whether the spin splitting of the subbands $e1$ or $lh1$ predominates, Ref. [29].

The injection of pure spin currents in (110)-oriented GaAs QWs at room temperature by one-photon absorption of a linearly polarized optical pulse was demonstrated by Zhao et al. [14]. Spatially resolved pump-probe technique was used which enabled the authors to obtain signatures of the pure spin currents by measuring the resulting spin separations of $1 \div 4$ nm. The pump pulse excited electrons from the valence to the conduction band with an excess energy of ~ 148 meV. The probe was tuned near the band edge. It was observed that the spin current resulting in separation of the spin density $S_{z'}$ along the $[1\bar{1}0]$ axis reversed its direction when the polarization of the pump pulse was switched from $\mathbf{e}\|x'$ to $\mathbf{e}\|y'$. This indicates that, for the photon energy used in the experiment, the polarization-dependent contribution dominates over the polarization-independent term.

IV. INTERSUBBAND TRANSITIONS IN N-DOPED QWS

The intersubband light absorption in n-doped QW structures is a resonant process which becomes possible if the photon energy $\hbar\omega$ is tuned to the intersubband energy separation. In the simple one-band model, direct optical transitions between the electron subbands $e1$ and $e2$ conserve spin and are induced only by radiation with a nonzero normal component $e_\perp$ of the polarization vector. If the spin degeneracy of the quantum subbands is lifted, such spin-conserving optical transitions give rise to a pure spin current [13, 30]. This mechanism is illustrated in Fig. 2, where the intersubband transitions $(e1, +1/2) \to (e2, +1/2)$ and $(e1, -1/2) \to (e2, -1/2)$ are shown by vertical solid lines. Due to **k**-linear spin splitting of the subbands together with the energy and quasi-momentum conservation, the optical transitions induced by light of a fixed frequency occur only at certain values of $k_{x'}$, denoted by $k_{+1/2}$ and $k_{-1/2}$ for the spin states $\pm 1/2$, respectively, where the photon energy $\hbar\omega$ matches the energy spacing between the subbands. As is evident from Fig. 2(a), these $k_{x'}$-points are of opposite signs for transitions from the spin branches $\pm 1/2$. Similarly to the interband light absorption considered in Sect. III, such spin-dependent asymmetry of photoexcitation gives rise to pure spin currents in both $e1$ and $e2$ subbands.

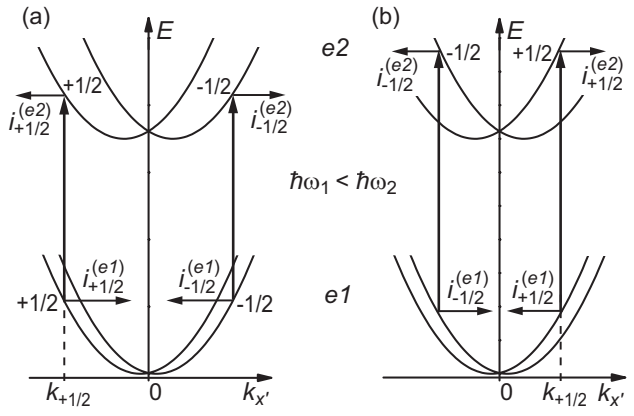

FIG. 2: Microscopic mechanism of the pure spin photocurrent induced by intersubband excitation with linearly polarized light due to **k**-linear splitting of the subbands. Panels (a) and (b) demonstrate the spin current reversal with increasing the light frequency.

An interesting feature of the pure spin photocurrent caused by **k**-linear splitting of the subbands is its spectral response. Figures 2(a) and 2(b) show what happens if the photon energy $\hbar\omega$ crosses the resonance varying from $\hbar\omega < E_{21}$ to $\hbar\omega > E_{21}$, where E_{21} is the energy separation between the subbands at $\mathbf{k} = 0$. For the photon energy below E_{21} [see Fig. 2(a)], the optical transitions $(e1, +1/2) \to (e2, +1/2)$ occur at negative values of $k_{x'}$ leading to a flow of spin-up electrons in the subband $e1$ in the x' direction. With increasing the light frequency, the point of optical transitions $k_{x'} = k_{+1/2}$ at which the energy and quasi-momentum conservation laws are met moves toward positive values of $k_{x'}$ [see Fig. 2(b)]. This results in an inversion of the spin current.

The explicit spectral dependence of the spin photocurrent in an ideal QW drastically depends on the fine structure of the energy spectrum. In real QW structures, the spectral width of the intersubband resonance is substantially broadened. Allowance for the broadening can be made assuming, e.g., that the energy separation E_{21} between the subbands varies in the QW plane [31, 32]. Then, to the first order in the spin-orbit coupling, the spin current components in the subbands are given by

$$J_\beta^{\alpha(e1)} = \frac{\tau_{e1}\, e_\perp^2}{2\hbar} \left(\gamma_{\alpha\beta}^{(e2)} - \gamma_{\alpha\beta}^{(e1)}\right) \bar{E}\, \frac{d\,\eta_{21}(\hbar\omega)}{d\,\hbar\omega} \frac{I}{\hbar\omega}\,, \quad (16)$$

$$J_\beta^{\alpha(e2)} = \frac{\tau_{e2}\, e_\perp^2}{2\hbar} \left(\gamma_{\alpha\beta}^{(e2)} - \gamma_{\alpha\beta}^{(e1)}\right) \left[\eta_{21}(\hbar\omega) - \tau_{e2}\bar{E}\, \frac{d\,\eta_{21}(\hbar\omega)}{d\,\hbar\omega}\right] \frac{I}{\hbar\omega}\,, \quad (17)$$

where τ_{e1} and τ_{e2} are the spin current relaxation times in the subbands $e1$ and $e2$, respectively, $\gamma_{\alpha\beta}^{(e1)}$ and $\gamma_{\alpha\beta}^{(e2)}$ are the constants of **k**-linear spin-orbit coupling in the subbands, see Eq. (14), $\eta_{21}(\hbar\omega)$ is the intersubband absorbance for radiation polarized along the QW normal with the inhomogeneous broadening being taken into account, and $\bar{E}$ is the mean value of the electron kinetic energy. The energy $\bar{E}$ equals to $E_F/2$ for a two-dimensional degenerate gas with the Fermi energy E_F and k_BT for a non-degenerate gas at the temperature T.

The spin photocurrents (16) and (17) are contributed by spin-conserving optical transitions and, therefore, are proportional to the difference of subband splitting constants. The spectral behavior of the pure spin currents in

both subbands repeats the derivative of the light absorption spectrum $d\eta_{21}(\hbar\omega)/\hbar\omega$ provided the intersubband absorption line is narrow enough. Close to the absorption maximum the spin photocurrents reverse their directions with varying the light frequency. We also note that the contribution $J_\beta^{\alpha(e1)}$ can considerably exceed $J_\beta^{\alpha(e2)}$ since the relaxation time in the excited subband τ_{e2} may be quite short even at low temperatures due to the effective channel of relaxation by emission of an optical photon.

A contribution to the pure spin currents may also come from linear-in-**k** spin-dependent terms in the matrix elements of optical transitions. While in the one-band approximation the intersubband absorption can only be induced by the $e_\perp$ component of the polarization vector, in a multi-band model optical transitions between the electron subbands $e1$ and $e2$ are allowed for any polarization [33]. Moreover, **k**·**p** admixture of the valence-band and remote conduction-band states to the electron wave functions adds both spin-dependent [34] and **k**-linear terms to the matrix elements of the optical transitions. Taking into account these contributions, the 2×2 spin matrix M_{21} describing the intersubband transitions assumes the form

$$M_{21} = M_{21}^{(0)}\left(e_\perp + i\sum_{\alpha\beta}\lambda_{\alpha\beta}\,\sigma_\alpha e_\beta + i\sum_{\alpha\beta}\lambda'_{\alpha\beta}\,k_\alpha e_\beta + \sum_{\alpha\beta\gamma}\lambda''_{\alpha\beta\gamma}\sigma_\alpha k_\beta e_\gamma\right), \quad (18)$$

where $M_{21}^{(0)}$ is the matrix element calculated in the one-band approximation for radiation polarized along the QW normal. The tensor λ is responsible for the intersubband optical orientation of electron spins [34], while λ' and λ'' describe the optical alignment of electron momenta. Taking into account **k**-linear terms in Eq. (18), we derive the contributions to pure spin currents excited in the $e1$ and $e2$ subbands as

$$J_\beta^{\alpha(e1)} = \left(\sum_{\gamma\delta}\lambda_{\alpha\gamma}\lambda'_{\beta\delta}\,e_\gamma e_\delta - \sum_{\gamma}\lambda''_{\alpha\beta\gamma}\,e_\perp e_\gamma\right) \times\frac{\tau_{e1}\bar{E}}{\hbar}\,\frac{I\eta_{21}(\hbar\omega)}{\hbar\omega}\,, \quad (19)$$

$J_\beta^{\alpha(e2)} = -(\tau_{e2}/\tau_{e1})J_\beta^{\alpha(e1)}$. In contrast to Eq. (16), the spectral response of the contribution (19) repeats the light absorption spectrum.

V. FREE-CARRIER ABSORPTION IN N-DOPED QWS

The light absorption by free carriers, or Drude-like absorption, occurs in doped semiconductor structures when the photon energy $\hbar\omega$ is smaller than the band gap as well as the energy spacing between the subbands. Such an intrasubband excitation of carriers with linearly polarized light also gives rise to a pure spin current. However, in contrast to the direct transitions considered in Sects. III and IV, the subband spin splitting leads to no essential contribution to the spin current induced by intrasubband optical excitation. The more important contribution comes from the spin-dependent asymmetry of electron scattering [13, 35]. Indeed, the free-carrier absorption is always accompanied by electron scattering from acoustic or optical phonons, static defects, etc., because of the need for energy and momentum conservation. In systems with a spin-orbit interaction, processes involving change of the particle wave vector are spin dependent. In particular, in the QW structures the matrix element of electron scattering $V_{\mathbf{k}'\mathbf{k}}$ contains, in addition to the main contribution V_0, asymmetric spin-dependent terms [34, 36]

$$V_{\mathbf{k}'\mathbf{k}} = V_0 + \sum_{\alpha\beta} V_{\alpha\beta}\,\sigma_\alpha(k_\beta + k'_\beta)\,, \quad (20)$$

where $\mathbf{k}$ and $\mathbf{k}'$ are the initial and the scattered in-plane wave vectors, respectively. This leads in turn to **k**-linear spin-dependent contribution to the scattering rate, which is determined by the matrix element squared. Microscopically, such terms in the scattering rate originate from structure and/or bulk inversion asymmetries similar to **k**-linear Rashba and Dresselhaus spin splitting of the electron subbands.

Due to the spin-dependent asymmetry of scattering, electrons photoexcited from the subband bottom are scattered in preferred directions depending on their spin states. This is illustrated in Fig. 3(a), where the free-carrier absorption is shown as a combined two-stage process involving the electron-photon interaction (solid vertical lines) and the electron scattering (dashed horizontal lines). The scattering asymmetry is shown by dashed lines of different thicknesses: electrons with the spin $+1/2$ are preferably scattered into the states with $k_{x'} > 0$, while electrons with the spin $-1/2$ are predominantly scattered into the states with $k_{x'} < 0$. Obviously, such an asymmetry of photoexcitation in the **k** space leads to a pure spin current, where the spin-up and spin-down electrons counter flow and the charge current vanishes.

In the perturbation theory approach, the indirect optical transitions are treated as second-order virtual processes involving intermediate states. To the first order in spin-orbit interaction, the compound matrix element of the intrasubband transitions accompanied by the electron scattering from short-range potentials has the form [37]

$$M_{\mathbf{k}'\mathbf{k}} = \frac{eA}{c\,\omega m_e}\,\mathbf{e}\cdot(\mathbf{k}'-\mathbf{k})V_{\mathbf{k}'\mathbf{k}} - 2\frac{eA}{c\hbar}\sum_{\alpha\beta}V_{\alpha\beta}\,\sigma_\alpha e_\beta\,, \quad (21)$$

where e is the electron charge, A is the vector potential of the electromagnetic wave, and c is the light velocity.

The first term on the right-hand side of Eq. (21) describes transitions $(e1,\mathbf{k}) \to (e1,\mathbf{k}')$ with intermediate states in the conduction subband $e1$, the second term corresponds to the transitions via intermediate states in other bands. We assume that the electron scattering is elastic and consider the geometry of normal incidence of the light so that the polarization vector $\mathbf{e}$ lies in the (xy) plane. Then, the polarization dependencies of spin current components are given by

$$J_x^\alpha = -\frac{\tau_e}{\hbar}\left(\frac{\langle V_0 V_{\alpha x}\rangle}{\langle V_0^2\rangle}\,\frac{e_x^2-e_y^2}{2} + \frac{\langle V_0 V_{\alpha y}\rangle}{\langle V_0^2\rangle} e_x e_y\right) I\eta_{e1}\,. \tag{22}$$

Here, the angle brackets $\langle ...\rangle$ stand for averaging over the spacial distribution of scatterers and η_{e1} for the radiation absorbance in this spectral range. The components J_y^α can be obtained from Eq. (22) by the replacement $x \leftrightarrow y$.

Equation (22) shows that pure spin currents can be injected in QWs by the elastic-scattering-assisted photoexcitation with linearly polarized light but vanish for the normally-incident unpolarized radiation, when $\overline{e_x^2} = \overline{e_y^2} = 1/2$, $\overline{e_x e_y} = 0$. The nonzero components of the spin current are determined by the explicit form of the matrix element of scattering and the light polarization plane. In QWs grown on (110)-oriented substrates, the scattering rate contains the term proportional to $\langle V_0 V_{z'x'}\rangle\,\sigma_{z'}(k_{x'}+k'_{x'})$ giving rise to the components $J_{x'}^{z'} \propto (e_{x'}^2 - e_{y'}^2)$, $J_{y'}^{z'} \propto e_{x'}e_{y'}$, which are in accordance with the phenomenological equation (5). In (001)-grown structures, the nonzero coefficients are $\langle V_0 V_{xy}\rangle = -\langle V_0 V_{yx}\rangle$ and $\langle V_0 V_{xx}\rangle = -\langle V_0 V_{yy}\rangle$, and the normally-incident radiation can excite fluxes of the in-plane spin components only.

The pure spin current caused by the free-carrier absorption can be converted into an electric current by polarizing electron spins, e.g., by application of an external magnetic field, as was shown by Ganichev et al. [35], see also [8]. Indeed, in the case of intrasubband absorption, the fluxes of the spin-up and spin-down electrons, $\mathbf{i}_{+1/2}$ and $\mathbf{i}_{-1/2}$, are proportional to the electron densities in the spin subbands, $n_{+1/2}$ and $n_{-1/2}$, respectively. In a spin-polarized system, where $n_{+1/2} \neq n_{-1/2}$, the fluxes $\mathbf{i}_{+1/2}$ and $\mathbf{i}_{-1/2}$ do no longer compensate each other yielding a net electric current

$$j_\beta = 4e\sum_\alpha S_\alpha J_\beta^\alpha\,, \tag{23}$$

where $\mathbf{S}$ is the average electron spin with $|\mathbf{S}| = (1/2)|n_{+1/2}-n_{-1/2}|/(n_{+1/2}+n_{-1/2})$.

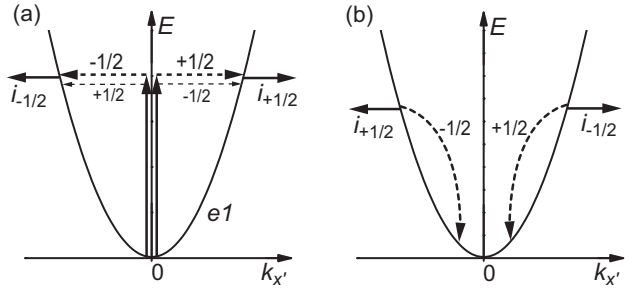

FIG. 3: Microscopic mechanisms of the pure spin currents caused by spin-dependent scattering processes at (a) intrasubband photoexcitation with linearly polarized light and (b) energy relaxation of hot carriers.

VI. PURE SPIN CURRENTS CAUSED BY ELECTRON GAS HEATING

In addition to the free-carrier absorption, the spin-dependent asymmetry of the electron scattering by phonons gives rise to a pure spin current if the electron gas is simply driven out of thermal equilibrium with the crystal lattice (see Refs. [8, 35, 38]). In such a relaxational mechanism, the spin current is generated in the process of energy relaxation of electrons no matter how the thermal equilibrium between the electron and phonon subsystems was initially disturbed.

The relaxational mechanism of the spin current generation is illustrated in Fig. 3(b), where the processes of energy relaxation of hot electrons by emitting phonons are shown by dashed curves. Due to the spin-dependent asymmetry of the electron-phonon interaction, electrons with the spin $+1/2$ relax faster from the high-energy states with positive $k_{x'}$, while electrons with the spin $-1/2$ predominantly vacate the high-energy states with negative $k_{x'}$. This leads to an asymmetrical distribution, where the spin-up carriers occupy mainly the left-hand branch of the dispersion curve (carriers with the opposite spin orientation have gone to the subband bottom), while the spin-down carriers occupy mainly the right-hand branch. Such a spin-dependent imbalance of electrons between positive and negative $k_{x'}$ yields a pure spin current.

We consider the energy relaxation of electrons confined in a QW by bulk acoustic phonons. Taking into account $\mathbf{k}$-linear contributions to the electron-phonon interaction, the matrix element of the electron scattering by phonons can be modeled by

$$V_{\mathbf{k}'\mathbf{k}}(\mathbf{q}) = \mathcal{V}_0(q_\perp) + \sum_{\alpha\beta}\mathcal{V}_{\alpha\beta}(q_\perp)\,\sigma_\alpha(k_\beta+k'_\beta)\,, \tag{24}$$

where $\mathcal{V}_0(q_\perp)$ and $\mathcal{V}_{\alpha\beta}(q_\perp)$ are functions of $q_\perp$ with their forms dependent of the QW design, and $\mathbf{q} = \pm(\mathbf{k}-\mathbf{k}', q_\perp)$ is the three-dimensional wave vector of the phonon involved. We assume that both electrons and phonons obey the Boltzmann statistics, but the electron temperature T_e differs from the lattice temperature T_0. Then, the rates of phonon emission and absorption become nonequal leading to a spin current

$$J_\beta^\alpha = -\frac{N_e}{2}\frac{\tau_e}{\tau_{ph}}\frac{\hbar\, c_s^2}{k_B T_0}\frac{T_e - T_0}{T_e}\times \tag{25}$$

$$\times \frac{\int\limits_{-\infty}^{+\infty} \mathrm{Re}[\mathcal{V}_0^*(q_\perp)\mathcal{V}_{\alpha\beta}(q_\perp)]|q_\perp|dq_\perp}{\int\limits_{-\infty}^{+\infty} |\mathcal{V}_0(q_\perp)|^2/|q_\perp|\,dq_\perp},$$

where N_e is the carrier density, τ_{ph} is the momentum relaxation time governed by the electron-phonon interaction, and c_s is the sound velocity in the crystal.

As a more detailed example, we consider the (110)-grown QWs. In this case the dominant spin-dependent contribution to the Hamiltonian of electron-phonon interaction in the deformation-potential model is proportional to $\sigma_{z'}(k_{x'}+k'_{x'})$, and the corresponding Hamiltonian has the form [39]

$$H_{\mathrm{el-phon}}(\mathbf{k}',\mathbf{k}) = \Xi_c \sum_\alpha u_{\alpha\alpha} + \xi\,\Xi_{cv}\,u_{z'z'}\sigma_{z'}(k_{x'}+k'_{x'})/2. \tag{26}$$

Here, Ξ_c and Ξ_{cv} are the intraband and interband constants of the deformation potential, $u_{\alpha\beta}$ are the phonon-induced strain tensor components, $\xi = P\Delta_{\mathrm{so}}/[3E_g(E_g+\Delta_{\mathrm{so}})]$, and Δ_{so} is the spin-orbit splitting of the valence band. The interband constant Ξ_{cv} originates from the lack of an inversion center in zinc-blende-type crystals and vanishes in centrosymmetric semiconductors [6]. Assuming that electrons are confined in a rectangular quantum well of the width a, we derive for the spin current

$$J^{z'}_{x'} = -\frac{\pi^2\xi}{3a^2}\frac{\tau_e}{\tau_{ph}}\frac{\hbar\,c_s^2}{k_B T_0}\frac{\Xi_{cv}}{\Xi_c}\frac{T_e-T_0}{T_e}N_e\,. \tag{27}$$

Equation (27) shows that the spin current component $J^{z'}_{x'}$ strongly depends on the QW width.

VII. PURE VALLEY-ORBIT CURRENTS

In addition to the spin, free carriers in solid states can be characterized by another internal property, e.g., by a well number in multiple QW structures or a valley index ν in many-valley semiconductors. In the latter case, one can consider pure orbit-valley currents, where partial electron fluxes in valleys $\mathbf{i}_\nu$ are nonzero but the net electric current $e\sum_\nu \mathbf{i}_\nu$ vanishes [13]. Here, the role of spin-up and spin-down states is replaced by the valley index: there is no net charge current, but the electrons in different valleys travel in different directions.

To elaborate the concept of pure orbit-valley currents, we consider silicon-based quantum wells grown on a (111)-oriented surface. In Si QWs, the conduction-band subbands are formed by six equivalent valleys X, X', Y, Y', Z, and Z' located at the Δ points of the Brillouin zone of the bulk crystal. All the valleys retain their equivalence in (111)-grown structures because the angles between the growth direction and the valley principle axes are the same. Figure 4 sketches the valley positions and orientations in the two-dimensional $\mathbf{k}$ space in the QW plane. In asymmetrical (111)-grown QWs, each valley has the C_s point-group symmetry allowing for the generation of a partial in-plane flux $\mathbf{i}_\nu$ at normal incident

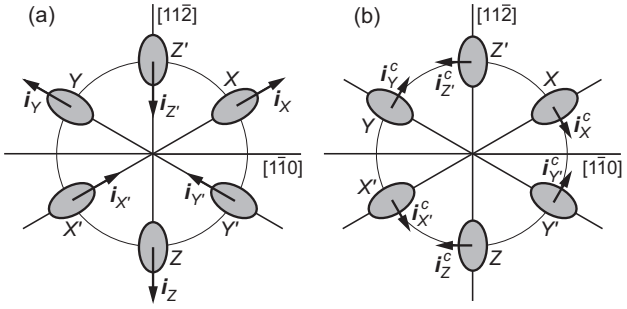

FIG. 4: (a) Angular distribution of electron fluxes in valleys under excitation with unpolarized light. (b) Angular distribution of helicity-dependent fluxes in valleys.

of the light. Under excitation with unpolarized light, the fluxes $\mathbf{i}_\nu$ are directed along the in-plane projections of the valley principle axes [see Fig. 4(a)]. Since the structure is invariant with respect to the rotation by 120° along the growth direction, the total charge current $e\sum_\nu \mathbf{i}_\nu$ vanishes. Thus, such an electron distribution can be referred to as an optically injected pure valley-orbit current.

In addition to the polarization-independent photocurrent, the excitation of a Si (111)-grown QW with circularly polarized light at normal incidence results, in each valley, in a flux component $\mathbf{i}^c_\nu$ which reverses its direction upon switching the light polarization from right-handed to left-handed circular polarization. Such helicity-dependent components $\mathbf{i}^c_\nu$ flow perpendicularly to the valley principle axes [see Fig. 4(b)] and also contribute to the pure valley-orbit current. We note that the absence of a total photocurrent under illumination with unpolarized or circularly polarized light is related to the overall C_{3v} symmetry of the QWs which, however, allows for a net electric current induced by linearly polarized light. In this particular case, the partial fluxes in valleys become nonequal and do not compensate each other.

Pure valley-orbit currents can also be optically injected in bulk multi-valley noncentrosymmetrical crystals such as AlAs, AlSb, GaP, etc. In these compounds, the conduction-band minima are located in the X points at the Brillouin-zone edge. Each of three equivalent valleys $\nu = X, Y, Z$ has the D_{2d} symmetry allowing for the helicity-dependent electron flux $\mathbf{i}_\nu$

$$\begin{aligned} \mathbf{i}_X &= \mathcal{P}I(0, æ_y, -æ_z)\,, \\ \mathbf{i}_Y &= \mathcal{P}I(-æ_x, 0, æ_z)\,, \\ \mathbf{i}_Z &= \mathcal{P}I(æ_x, -æ_y, 0)\,, \end{aligned} \tag{28}$$

where $æ_\alpha$ ($\alpha = x, y, z$) are components of the vector $\mathrm{i}[\mathbf{e}\times\mathbf{e}^*] = P_{circ}\,\mathbf{q}/q$, $\mathbf{q}$ is the light wave vector, and P_{circ} is the light helicity ranging from -1 to $+1$. In accordance with the overall T_d point group of the zinc-blende-type crystals, the total current vanishes for the homogenous illumination with circularly polarized light. In an external magnetic field, each contribution $\mathbf{i}_\nu$ varies due to the Lorentz force acting upon electrons. This action is,

however, different for different valleys due to the energy spectrum anisotropy in valleys. As a result, the magnetic field causes an imbalance of the valley-orbit current giving rise to a nonzero net electric current

$$\mathbf{j} \propto (æ_y B_z + æ_z B_y, æ_z B_x + æ_x B_z, æ_x B_y + æ_y B_x)\,.$$

For the particular geometry $\mathbf{q}\|[111]$ and $\mathbf{B}\|[010]$, the magnetic field induced photocurrent $\mathbf{j}$ appears in the [101] direction.

VIII. CONCLUSION

We have shown that pure spin currents of free carriers can readily be created in semiconductor structures by optical excitation with linearly polarized or even unpolarized light. The pure spin currents lead to spacial separation of the spin-up and spin-down particles and accumulation of the opposite spins at the opposite edges of the sample. We have presented the microscopic theory of pure spin photocurrents for all main types of optical transitions ranging from the fundamental interband to the free-carrier absorption. In the present paper we have focused on the spin photocurrents contributed by charge carriers. In addition, spin fluxes (or, in general, angular-momentum fluxes) can also be formed by neutral particles or excitations lacking electric charge such as photons [40, 41], excitons or exciton polaritons [42, 43], and even phonons and magnons. The study of spin currents is naturally inscribed in the physics of spin-related phenomena and opens up new opportunities for the realization of novel device concepts.

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